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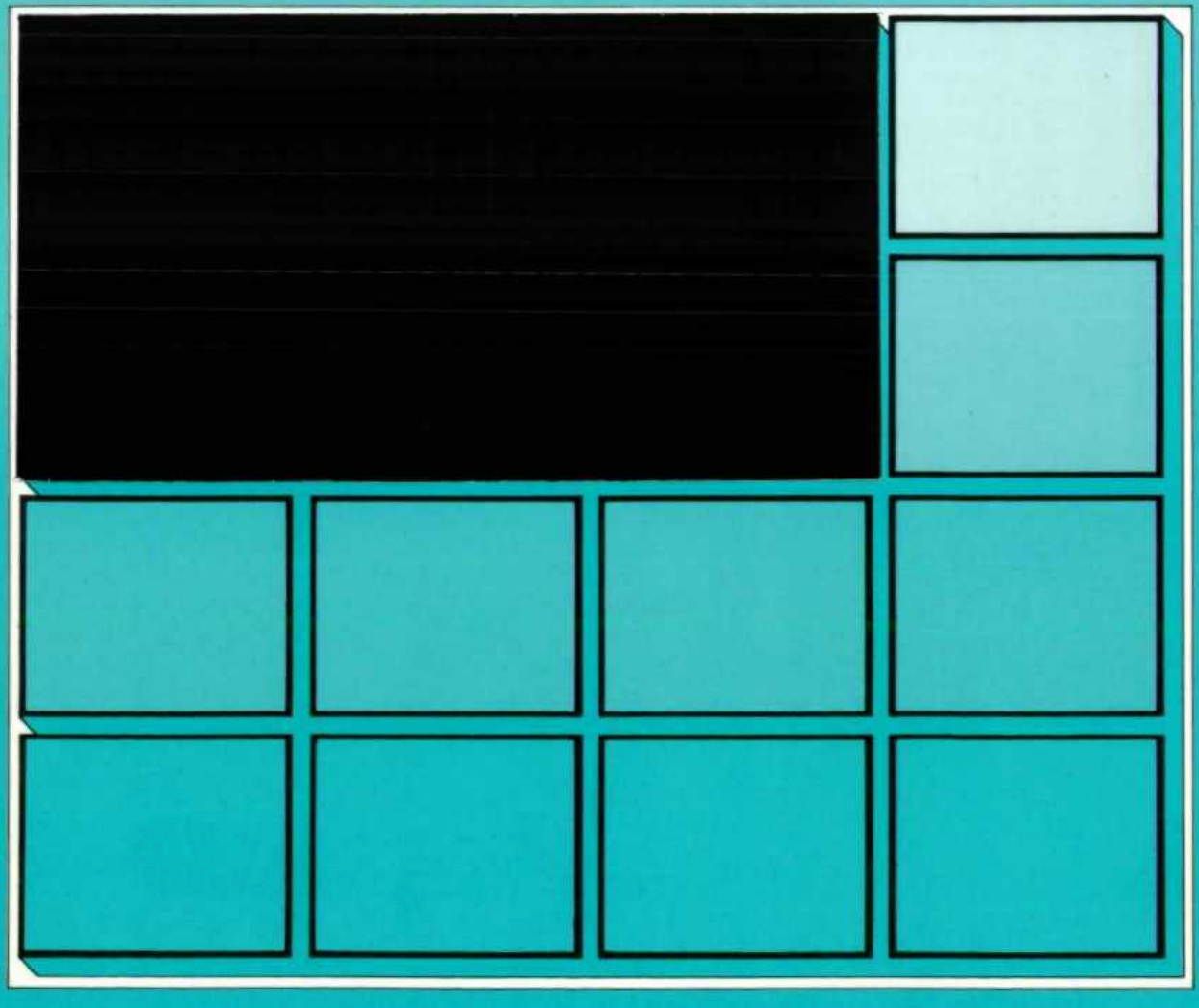

Modelling Caesium Transport

Summaries and abstracts of selected literature

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Abstract

The purpose of this review is to indicate the present level of understanding of the various processes that influence the transport of atmospherically dispersed caesium, through catchments and aquatic systems into freshwater fish. The areas investigated include deposition, soil adsorption, water chemistry, sedimentation and biological uptake.

The literature tends to suggest that for caesium deposition the most important process is wash-out, since Cs concentrations on grass and in soil are highly correlated with rainfall. Furthermore in the case of Chernobyl, the deposition occurred over a relatively short period of time, as, unlike the weapons fallout the Chernobyl radionuclides were only dispersed into the troposphere, and correspondingly had a shorter residence time, about a week in the atmosphere.

When deposited up to 30% of the Cs is intercepted by plants. This is the main route for contamination of terrestrial foodstuffs. The remainder of the caesium is rapidly and efficiently adsorbed by the soil. This adsorption is partly by fixation, and partly mediated by cation exchange sites. As a result of this strong adsorption there is very little wash off or downward movement of the radionuclide.

The wash off factor for freshly deposited fallout caesium is estimated by various authors, values range form 0.1 to 30 %, but most are between 1 and 5 % an average value being about 2%. Wash off of cumulated deposit is also estimated, values range between 0.1 and 1 %, with the average being about 0.6%. Thus the most important process by which caesium is lost from the soil is physical decay ($t_{1/2}$=30 years).

There is also very little movement down through the soil profile, although it does appear that there is far greater mobility in peats, possibly as a result of the far lower concentrations of illite and other micaceous minerals. Thus the contamination of groundwater by the leaching of atmospherically deposited caesium is unlikely, although contamination of groundwater has occurred in the U.S. as a result of leakage from underground waste depositories.

The caesium entering water bodies by direct deposition is in soluble form, and much of it is probably washed out of the water body in this form. Most of the remainder is adsorbed onto suspended sediment whilst still in the main body of water, or it is adsorbed onto bed sediment by the mechanism of diffusion into interstitial water and thence onto the sediment. This adsorption process is of such efficiency that the ratio of water concentrations to sediment concentrations is of the order of 1:30.

Introduction

Caesium is an alkali metal, atomic no. 55. It is soluble in water were it exists in the form Cs^+. Only the fluoride of Cs^+ is noticeably insoluble. The radionuclide appears to form soluble phases in rainwater, sometimes hydrated, except when fixed.

Although there are no direct results of comparison between the behaviour of Cs^+ in weapons test fallout and that in nuclear accident releases, it would appear reasonable to assume similarity in behaviour after deposition since caesium rapidly undergoes exchange and is sorbed onto soil materials.

Mobility -	in oxidising	acid	neutral/alkaline	reducing conditions
	low	low	very low	low/immobile

It is demonstrated that ^{137}Cs in microquantities is not present on normal cation exchange sites of soils, but is rather selectively sorbed at micaceous sites. In larger quantities the caesium is partly adsorbed onto exchange sites before migration to non-exchangeable sites. This leaves some caesium available for uptake by plants. Once the caesium is fixed there is very little evidence of movement, it is rarely desorbed from soils, although it has been observed to desorb from estuarine sediments in conditions of high salinity, as would be expected given its conservative behaviour in seawater.

Radiocaesium is mainly produced by nuclear fission reactions, although there is a small amount present in naturally occurring minerals(Thornton 1983). Cs-137 accounts for about 6% of all the fission products of uranium.. Once produced it then decays itself according to the following scheme

$$^{137}Cs \xrightarrow[t\frac{1}{2} = 30\ yr]{\beta^-} {}^{137}Ba \qquad \text{Strong Beta emitter}$$

$$\lambda = 0.034\ yr^{-1}$$

The nature of the fission reaction which produced the caesium can be elucidated by the ratios of it to other radionuclides. For instance there was very little Cs-134 in the weapons tests fallout of the 1960s, whilst the ratio of Cs-134 to Cs-137 in Chernobyl fallout was about 0.6 : 1. Thus it has been possible by analysis of samples (using Germanium drift beta detectors) to estimate the quantities of caesium present and its origin. Also quite importantly, the ratio of Cs to strontium in various environmental compartments has been measured, allowing comparison of the transport behaviour of caesium with that for the better known strontium. The ratio of Cs to Sr in rainfall after the Chernobyl incident was about 1.6.

1. Meteorology

1.1 FALLOUT IN AIR AND RAIN

1.1.1 Weapons Tests Monitoring

Data from AERE monitoring stations throughout the U.K. give figures for fallout in air and rain from 1954 onwards (Cambray 1964-). The figures for the early 1960's (1964) give values of 10-50 p Ci/litre for the total activity deposited in rainfall. This value drops by about 40% for each of the subsequent years when no testing was carried out. In 1969 and 1970 this decrease slows and there was an increase in deposition due to a Chinese weapons test.

Subsequently concentrations of Cs-137 in rainfall continue to decrease until in 1973 activity is less than 2% of the maximum of 1963/64.

Figure 1 shows the of cumulative deposition of Cs-137 (Cambray *et al.* 1987) and it is apparent that until the Chernobyl accident of 1986 there was little significant deposition of Cs-137 since the weapons testing of the 1960's.

It is also apparent from Tables 1 and 2 (Cambray et al., 1987) that the fallout from Chernobyl was mostly deposited in the period of May and June 1986. This rapid fallout is a result of the lower height that the debris from Chernobyl was emitted in comparison with the weapons tests of the 1960's. It is estimated that debris was still being emitted from the reactor at Chernobyl until it was encased in concrete in July (?) 1986.

1.1.2 Chernobyl

The radioactive cloud produced by the chernobyl accident hovered between 1000 - 3000 m above sea level brushing high ground in the western parts of the north of England, Scotland and Wales. Provincial rainfall caused great variations in the deposition of radioactivity, with local spots sometimes showing 50 times average values even within areas covered by the same plume.

Initially (28-29 April 1986) the concentrations of Cs-137 remained constant at 2.5 Bq m^{-3} in air. On 2 May only small concentrations were observed, although on the 8 May quite high concentrations, comparable to those on 28-29 April were observed before they declined again by 2 orders of magnitude (Derell, 1986).

It is clear from the observed concentrations of radionuclides that the volatile nuclides (^{131}I, ^{137}Cs, ^{136}Te, ^{90}Sr) are more dominant in the fallout than non-volatile elements (^{103}Ru, ^{141}Ce, ^{239}Np).

Caesium activities for air and rainfall are given for May 1986 at various locations are given in Cambray (1987)

Figure 1 *Cumulative deposition of caesium-137 (from Cambray et al. 1987)*

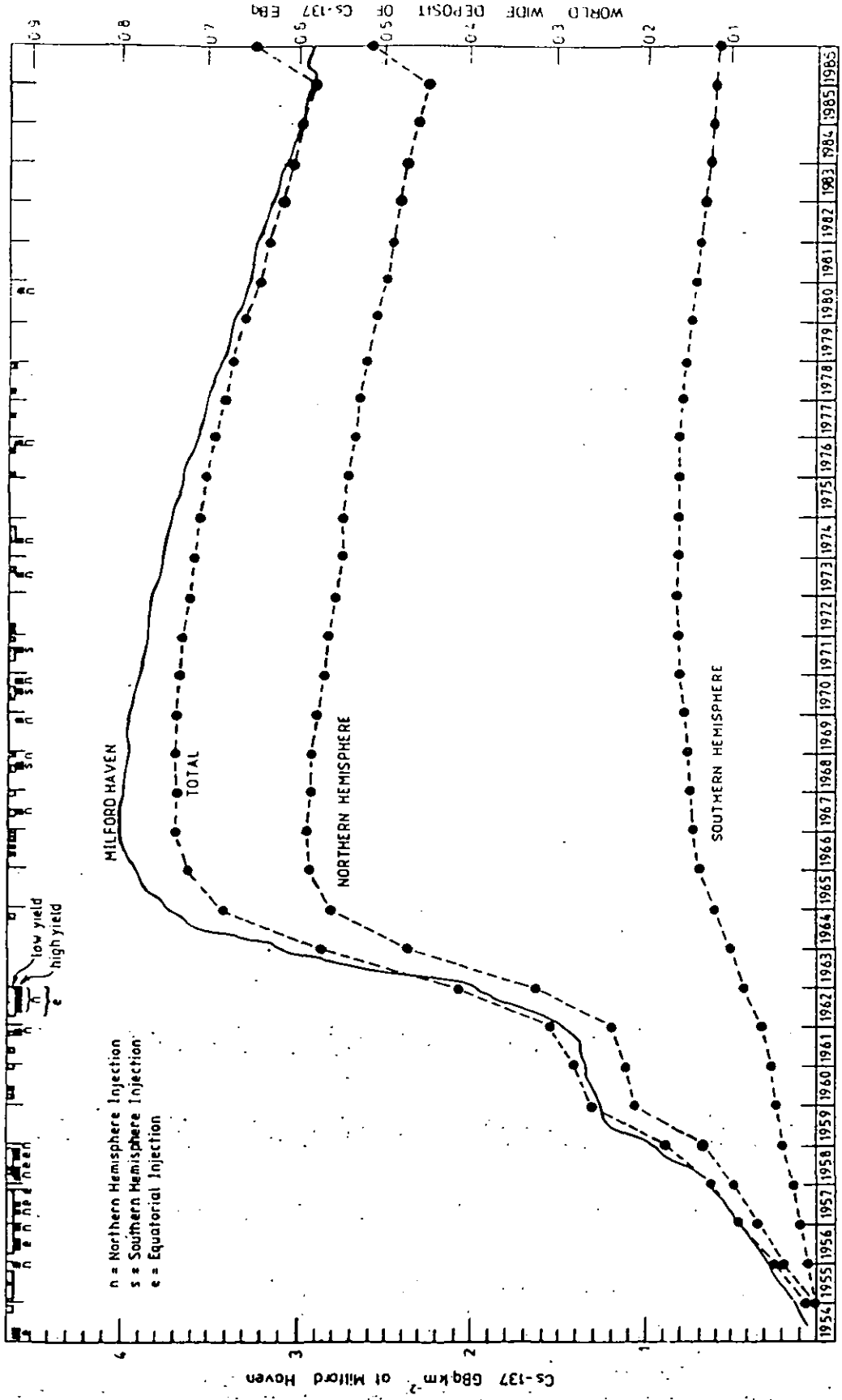


Table 1 Quarterly concentration of Cs-137 in rain. mBq.l^{-1}

Station	1985 1	2	3	4	1986 1	2	3	4
Lerwick	<20	<10	<10	<10	<10	30000	260	<20
Glasgow	-	-	-	-	-	28000	500	200
Eskdalemuir	<10	<10	<5	-	<15	1700	52	35
Aldergrove	<10	<10	<10	<25	<10	4200	350	290
Conlig	-	-	-	-	<10	12500	490	79
Snowdon	<25	<10	<15	<10	<20	9100	510	24
Orfordness	<25	<20	<30	N.S.	N.S.	1450	53	<25
Milford Haven	<10	<30	<10	<5	<25	160	29	8
Chilton	<20	<10	<5	<15	<20	330	73	<20
Compton	<15	<15	<15	<10	-	810	90	<20
London	-	-	-	-	-	450	200	-
Tromso	<15	<30	<10	<5	<50	960	1500	270
Reykjavik	<5	<35	<20	<5	<15	155	<50	<10
Esquimalt	-25	<15	<5	-	<25	75	410	14
Ottawa	<15	<10	<15	<10	<40	71	<10	<10
Gibraltar	-	<150	N.R.	<10	18	N.R.	270	46
Akrotiri	<10	N.R.	N.R.	<25	<15	3900	1150	350
Nassau	-	N.S.	N.S.	N.S.	N.S.	74	23	<3
Hong Kong	<5	<5	<5	<10	<20	120	6.9	<5
Singapore	<10	<20	5	<5	<10	S.L.	<6	-
Pretoria	<1000	<1000	<120	<20	<35	<35	<200	<5
Gaborone	-	-	-	-	<150	<200	<500	<15
Brisbane	<5	<10	<5	<5	<15	<25	<50	<5
Ohakea	<5	18	<5	-	<10	<10	<10	<10
Argentine Is	<20	<30	<20	<25	<10	<5	<5	<15
Halley Bay	<10	<15	<20	<15	<15	<5	<10	<15

S.L. = Sample Lost
N.R. = No Rain
N.S. = No Sample

"less than" results represent 1σ on the background.

1 millibecquerel = 10^{-3} Bq = 27 femtocuries

Table 2 Quarterly Deposition of Cs-137 by Rain. $Bq.m^{-2}$

Station	1985 1	2	3	4	1986 1	2	3	4
Lerwick	<6	<2.5	<2.5	<3	<3	5400	75	<10
Glasgow	-	-	-	-	-	9100	93	92
Eskdalemuir	<2.5	<2	<4	-	<6	760	15.5	28
Aldergrove	<1	<2	<2.5	<4	<2	980	60	75
Conlig	-	-	-	-	<2	2500	90	14
Snowdon	<25	<10	<15	<10	<20	7300	350	27
Orfordness	<2.5	<4	<6	N. S.	N.S.	100	3.5	<5
Milford Haven	<2	<5	<2.5	<1	<3	36	6	<4
Chilton	<3	<2.5	<1	<2.5	<3	53	14	<5
Compton	<2	<3.5	<2.5	<2	-	105	<10	<4
London	-	-	-	-	-	30	20	-
Tromso	<2.5	<5	<2	<1.5	<10	175	250	41
Reykjavik	<1.5	<4	<2	<2	<5	31	<10	<2.5
Esquimalt	4.8	<1.5	<1	-	<6	3.7	16	2
Ottawa	<1.5	<2.5	<2.5	<1	<4	19	<2	<1.5
Gibraltar	-	<5	N.R.	<3	1.4	N.R.	17	4.6
Akrotiri	<1.5	N.R.	N.R.	<2.5	<2	50	38	29
Nassau	-	N.S.	N.S.	N.S.	N.S.	13	7	<1
Hong Kong	<1	<4	<2.5	<1	<2.5	140	75	<1
Singapore	<2	<5	<2	<4	<7	S.L.	<3.5	-
Pretoria	<30	<25	<5	<50	<5	<3	<2	<5
Gaborone	-	-	-	-	<5	<10	<15	<15
Brisbane	<1.5	<2	<1.5	<1.5	<2.5	<3	<15	<2
Ohakea	<0.5	1.7	<0.5	-	<2.5	<2.5	<1.5	1.1

S.L. = Sample Lost
N.R. = No Rain
N.S. = No Sample

"less than" results represent 1σ on the background.

1 becquerel = 27 picocuries

1.1.3 Deposition

For Chernobyl the observed deposition velocities were as expected, but an order of magnitude lower than those normally used in accident studies as most of the larger fallout particles had been deposited before the plume reached the U.K..

Dry deposition velocity;

$$Vg = \frac{\text{quantity deposited per unit area of ground or surface}}{\text{mean concentrations in air x time}}$$

For grass, Cs-137, Cs-134 have calculated Vg of 0.5 mm s^{-1}. This is dependent on geometry of surface, airspeed and particle size.

Deposition in Rain;

Two processes

Rain out: radionuclide absorbed into cloud mass
Wash-out: precipitation scavenging - particles distributed in falling rain.

Wash-out factor;

$$W = \frac{\text{activity per unit mass of rain}}{\text{activity per unit mass of air}}$$

The wash-out factor for Cs isotopes, ^{131}I and ^{140}La are similar, being initially of the order of several thousand, but not exceeding a few hundred in the second half of the month (Garland, J.A.. and Cambray R.S.).

Where rainfall coincided with the plume line, Chernobyl deposition was two or three orders of magnitude greater than in dry areas. Observations confirm the overwhelming predominance of precipitation scavenging over dry deposition for small particles (Clark, 1988)

The concentrations of Cs^{137} in the air measured at Harwell and at several other stations in the UK are consistent with the hypothesis that by 20 May the Cs-137 had become distributed rather uniformly throughout a well defined volume of the atmosphere, and that subsequently concentrations remained rather uniform and removal could be described as a high order process with constant rate coefficient. The mean residence time of the radioactive materials in the tropopause of the Northern Hemisphere was $\simeq$ 7 days (Garland and Combray).

1.2 RESUSPENSION

After July 1986 the airborne concentrations of ^{137}Cs and the deposition in rain decreased much more slowly, and large variations have occurred between stations.

The sustained air concentrations could result from;

(i) the return of some activity into the lower stratosphere;

(ii) the resuspension and dispersal on a continental scale of material deposited near the reactor site;

(iii) local resuspension.

A further possibility, the resuspension in sea spray of activity deposited in the sea, can readily be shown to be incapable of contributing significantly. Local resuspension seems necessary to explain many features of the variation of concentrations in space and time. Most puzzling, however, is that a large fraction of material would need to be resuspended to explain the observations. Within a few weeks of the accident, only a few per cent of the activity on grassland remained on the vegetation. Most was found in the surface soil and a few per cent had penetrated to 3 or 4 cm depth. Apparently some mm of soil would be required in the deposition gauge to explain the collected ^{137}Cs, and such quantities are not observed in the gauges.

A study of resuspension from grass and soil surfaces at Harwell has shown that a resuspension factor of 10^{-6} m^{-1} applies for a few months after a deposit of large (> 8 μm) insoluble particles in the conditions around Chilton. Resuspension factors estimated from the concentrations of Cs-137 in air for the 3rd quarter of 1986 and the local deposit from the Chernobyl accident gives values varying from about 10^{-9} m^{-1} at Lerwick to about 5×10^{-8} m^{-1} at Chilton and Milford Haven and 1.7×10^{-7} at Orfordness. The larger estimates for the resuspension factors generally apply to areas where the deposit is low, and this suggests there is aerial transport from regions of high deposit.

1.2.1 Terrestrial

In the resuspension process surface particles become airborne due to physical disturbances, eg: rain, wind, animals, man. The magnitude and time dependence of the process is dictated by the nature of the surface and of the disturbance. Resuspension of radionuclides can occur long after their original deposition, and is of most importance for long-lived radionuclides especially those which are less readily transferred through food chains.

Compared with undisturbed surfaces, ploughing and cultivation of land on which deposition has occurred is expected to reduce the initial wind-driven resuspended air concentrations because of dilution of the contaminated surface layer, although in the long term it may increase if by returning contamination to the surface.

The resuspended air concentration is observed to decline with time after the material is first deposited. The decline is due to the surface deposit becoming progressively less available for resuspension as a result of chemical and physical changes.

$$K = \text{resuspension factor } m^{-1} = \frac{\text{Resuspended air concentration (Bq } m^{-3})}{\text{Surface deposition (Bq } m^{-2})}$$

Data suggest K declines from values in the range of 10^{-4} to 10^{-6} m^{-1} shortly after deposition to 10^{-8} - 10^{-9} m^{-1} after a few years (Garland).

One resuspension model (NRPB,CEA: CEC July 1979) is based on the relationship

$$K = 10^{-5}(\exp(-(\lambda_1 + \lambda_2 + \lambda)t)) + 10^{-9} \exp(-(e^{+} + \lambda)b)$$

K = resuspension factor m^{-1}
t = time after initial deposition (s)
λ_1 = decay constant for initial decline of K
λ_2 = decay constant for longer term decline of K
λ = radioactive decay constant of the nuclide of interest

Values used in a model

$\lambda_1 = 1.46 \times 10^{-7}$ s^{-1} half-life 0.25 years

$\lambda_2 = 2.2 \times 10^{-10}$ s^{-1} half-life 100 years

1.2.2 Maritime

From a study in Cumbria (Cambray 1982) it appears that Pu and to a lesser extent An are being resuspended from the sea, and are enriched in the spray; whilst Cs-137 shows no significant enrichment. The apparent existence of an enrichment effect for Pu and An in sea spray, and its absence for G-174 suggests that the mechanism may be related to the sediment in seawater rather than to material in solution.

2. Fluvial processes

2.1 RUNOFF

Upon deposition a portion of the fallout Cs-137 is retained on foliage, this portion may vary but for grass is 0.05 - 0.30. The loss of ^{137}Cs from the grass can be described by a simple exponential process with a half-life of about 14 days.

Subsequently the Cs is absorbed by the soil. By 1986 the Cs-137 remaining

from weapon tests was 200 Bq m^2 per 1000 mm annual rainfall, 75% of which was retained in the top 15 cm of soil. Run-off accounts, therefore, for only a small proportion of the caesium.

Values from Japanese rivers imply that 1.3% of Cs-137 and 7.2% of Sr-90 from the land surface which originated from fallout would be washed into rivers (Yamagata 1963). This is corroborated by figures given by (Carlsson 1978) who stated in his study of a small watershed in Sweden that only 1.9% of the freshly deposited Cs^{137} and 0.56% per year of the accumulated Cs-137 in the drainage area of the lake is transported to the lake.

In his review Helton (1985) states that it has been found to be possible to relate fallout rates and accumulated fallout levels to radionuclide concentrations in various environmental compartments by relatively simple empirical relationships.
The simplest of these relationships is:

$$Ci = a\,(D)i + b\,A_{i-1(n)}$$

Where;

Ci is the concentration in year i (Ci/l or Ci/kg)

$(D)i$ = fallout rate ($Ci/km^2/yr$)

$A_{i-1(n)}$ = accumulated fallout with an assumed half life of n years (Ci/km)

For a given region, the coefficients a and b depend on the size of the region, the amounts of runoff from the region, and the functions of recently deposited and accumulated radionucide removed in runoff. In particular a and b are given by :

$$a = L\,\lambda_a/R \qquad b = L\,\lambda_b/R$$

Where:

L = area of watershed (km^2)

R = annual runoff rate (l/yr)

λ_a = fraction of radionuclide (recently deposited) removed by runoff

λ_b = rate constant for the removal of accumulated radionuclides (yr^{-1})

The fact that initial wash-off rates are generally higher than subsequent wash-off rates is probably due to time dependent processes such as the movement of radionuclides into the soil column and the fixation of radionuclides to soil materials. The higher wash-off rates for smaller plots probably result from factors such as short-travel paths to collection points and use of experimental plots with little or no plant cover.

If x_0 (units : Ci) denotes the total amount of radionuclide with a radioactive

decay constant λ (units : yr^{-1}) initially released to a land surface, and x(t) denotes the amount present on the land surface at time t. Then the change of x(t) subsequent to the initial wash-off is described by the differential equation

$$\frac{dx}{dt} = -(\lambda + \lambda b)x \qquad x(o) = (1 - \lambda a)x_o$$

Which has the solution;

$$x(t) = (1 - \lambda a)x_o \exp [- (\lambda + \lambda b)t]$$

There the total amount W (units:Ci) of the radionuclide washing into the surface-water body is given by:

$$W = \lambda_a x_o \int_o^\infty \lambda_b x (+) dt$$

$$= \lambda_a x_o + (1 - \lambda_a)\lambda_b x_o/(\lambda + \lambda_b)$$

$$= (\lambda_a \lambda + \lambda_b) x_o/(\lambda + \lambda_b)$$

From calculations using this model for strontium, and using Yamagata's (1963) figures for caesium Helton concludes that the contamination of surface water bodies by the erosion of atmospherically deposited radionuclides is not a major risk associated with nuclear accidents.

2.2 SEDIMENT

2.2.1 Adsorption

As stated in the introduction, and as will be elaborated in the following two sections, it seems clear from experimental evidence (Francis 1976) and from theoretical studies (Francis 1971, Schulz 1959) that caesium is preferentially adsorbed by micaceous minerals in both soils and freshwater sediments For example Francis (1976) found that Cs-137 is predominantly retained by the fractions of freshwater mineral sediment containing muscovite and kaolonite.

The adsorption of caesium onto these minerals is affected by a number of factors, which have been investigated by Elprince (1978) and Elprince *et al.* (1977).

The dependence of Kd* on temperature, pH and salinity.

Work by Elprince suggests that the distribution coefficient,

$$kd^* = \frac{\text{Cs adsorbed} \sim \quad (\text{meq } g^{-1})}{\text{Cs in solution} \quad (\text{meq } l^{-1})}$$

is both temperature and pH dependent such that;

$$\ln kd^* \propto \frac{1}{T} \quad \text{and} \quad \ln kd^* \propto pH$$

which is experimentally shown to be a result of the adsorption of Cs on hydroxy Al interlayers.

$$Kd^* \sim 20\text{-}150$$

ln Kd* also appeared to show a linear relationship with concentrations of Na^+ and K^+ which competes with Cs^+ for adsorption sites. This is corroborated by the observations of Linslata (1986) on the Hudson river estuary, where caesium was apparently desorbed from sediments as a result of higher than usual salinity caused by a very dry summer in 1980. There is also evidence from a study by Cambray (1982) that high salinity may inhibit the retention of Cs-137.

The following is a summary of a paper by Lerman (1971) which indicates the overriding importance of sediment in controlling the water concentration of caesium.

The amount of ^{137}Cs in sediment which comes from Lakes Ontario and Superior is 20-30 times greater than the amount of ^{90}Sr. These high concentrations of Cs-137, viewed against the value of 1.45 for the $^{137}Cs/^{90}Sr$ ratio in atmospheric fallout and 0.16-0.36 in the Great Lakes water, indicate that Great Lakes sediment strongly control the concentration of ^{137}Cs in lake water.

The values of the ratio $^{137}Cs/^{90}Sr$ (0.36, 0.24, 0.16) in the water of Lakes Superior, Huron, and Ontario are lower than the value of approximately 1.45 in the atmospheric fallout. The low values in lake water are compatible with the relatively large amounts of ^{137}Cs in the lake sediments.

$$\text{Cs concentration} \sim 0.1\text{-}0.5 \text{ pCi/l}$$

^{90}Sr and ^{137}Cs occur only within the upper 9-11cm of the sediment column. The details of the distribution of the radionuclides in sediment sampled every 1 cm, indicate that the mechanism of entry into the sediment is diffusion from lake water into interstitial water and uptake by solid particles in the sediment. The amount of sediment in the section 9-11 cm deep is greater than the amount deposited during the last 20 years, as can be estimated from the mean rates of sedimentation of 0.03-0.08 cm/yr in lakes. The latter argument strengthens the evidence against the entry of ^{90}Sr and ^{137}Cs into the sediment via settling particles only.

The amount of Cs-137 in the Lakes Superior and Ontario sediments is a factor of 20-35 times higher than the amount of ^{90}Sr. These high values are comparable to the ratios $^{137}Cs/^{90}Sr$ in Lake Michigan sediments, where

values in the range 3 to 43, with the mean value near 18, have been recorded in 15 sediment core samples.

For Cs-137 the amounts stored in the sediments of Lakes Superior and Ontario are much greater than the amounts in the lake water. Thus, in the case of Cs-137 the lake sediments apparently exact a much more effective control of the concentration in lake water than in the case of ^{90}Sr.

2.2.2 Transport

Cs in reservoir sediments appears to originate from fallout onto catchments and watersheds. The Cs is adsorbed by clays and organic matter in soils, and this is then eroded and deposited as sediment. McHenry *et al.* (1973) indicated that for various American watersheds that the content of Cs per unit area was greater in sampled sediments than in the contributory watersheds and the relative concentration factor was as large as 24. For 1963/64 the amount of Cs-137 measured in sediments was at least five times greater than that deposited from fallout. Walters *et al.* (1982) found generally higher levels of activity are associated with bed sediment than with suspended sediment. For both bed and suspended sediment the clay fractions had the highest activity levels overall followed by silt and then sand. Radionuclides were found much less frequently in the water than in either the bed sediment or suspended sediment samples. Lerman (1971) had already argued that three processes dominate the distribution profile of radionuclide concentrations in sediment:

Sedimentation rate	U	$\approx$ 0.05 cm yr^{-1}
Diffusion	D	$\approx$ 3 x 10 cm sec^{-1}
Decay	λ	(Cs half-life = 30 yrs) 0.023 yr^{-1}

For Cs this means that the amount of nuclide stored in sediment at steady state is of the order of 12-60 times greater than the amount in the lake water.

Pennington *et al.* (1973), and Eakins and Cambray (1985) tend to suggest there is little vertical movement of Cs-137 in sediments, and that, consequently, levels of Cs activity correlated with deposition data can be an indication of the age of sediment and thus the rate of sedimentation. There have been failures with this technique, most of which have been attributed to bioturbation.

3. Catchment studies

In a study by Eakins and Cambray (1985) of a Cumbrian lake, the following

data emerged. Accumulations of ^{137}Cs, ^{210}Pb and Pu in the lake represent 1.5, 4.1 and 2.4% respectively of that retained on the catchment. The radionuclide content of the lake water is generally less than 1% of that retained in the lake sediment. The results of the investigation suggest that the bulk of the ^{210}-Pb present in the lake sediment is transferred from the catchment in association with decaying organic matter. Cs-137 is probably transferred with clay minerals and plutonium has also been shown to be associated with suspended particulate matter.

At each of the five sampling sites, at least 70% of the Cs-137 and Pu was found in the top 5 cm of soil with 90% in the top 10 cm. The retention of Cs-137 by the top 15 cm of soil averaged 75% of the amount present to the 30 cm depth.

^{137}Cs was distributed uniformly throughout the depth of the lake. Cs appears to be at similar concentrations in lake, inlet and outlet waters. Cs^{137} is believed to be retained on the catchment by ion exchange on clay minerals and hence should be at higher concentrations in water when erosion occurs during high floods. Only 34% at most of the ^{137}Cs deposited in (1976-77) sediment during this period could have been derived from direct input via rain. However, the retention by sediment was only 8% of the calculated input and there is no reason to believe that Cs-137 deposited directly into the lake will be accumulated in sediment at a faster rate than that entering from the catchment, in fact the converse is probably true.

McHenry and Ritchie (1975) concluded similarly that once in contact with the soil Cs-137 becomes strongly adsorbed on the finer soil fractions, and further downward movement by physicochemical processes is limited. Therefore most of the redistribution of Cs-137 within watershed ecosystems should be related to the erosion cycle in the watershed. The authors indicated that considerable amounts of Cs-137 fallout were lost from cultivated areas. Similar losses were observed on cultivated portions of other watersheds. The losses may be ascribed to soil erosion, and perhaps to some removal of Cs-137 in harvested crops. Table 3 shows the distribution of fallout Cs-137 in Murphy watershed, Mississippi.

Data shows that most of the fallout Cs-137 falling on S Eastern watersheds is still on that watershed, undoubtedly adsorbed by the finer soil particles. Some Cs-137 has been removed from the watershed. A portion of this Cs-137 is found in the river sediments. On an area basis there has been an increase of Cs-137 in these reservoir sediments over that in the watershed soils.

Table 3 Distribution of fallout Cs-137 in Murphy watershed, Mississippi; (from McHenry and Ritchie, 1975)

Input from atmosphere into reservoir and watershed				
152.8 nCi/m^2		Input activity		
53.87 ha		Area		
82.31 mCi		Total Activity		
1.75mCi	62.46mCi	12.77mCi	2.49mCi	Input
Forest	Grass	Cultivated	Eroded	Soil & Litter
149.7nCi/m^2	131.6nCi/m^2	99.6nCi/m^2	4.9nCi/m^2	Measured Conc.n
11.5ha	40.88ha	8.36ha	1.63ha	Area
1.72mCi	53.80mCi	8.33mCi	0.08mCi	Total Activity
0.04mCi	8.66mCi	4.44mCi	2.41mCi	Loss
		15.55mCi		Total Input
		2.83mCi		Direct Input
		470.1nCi/m^2		
		1.85ha		Reservoir
		8.70mCi		
		9.68mCi	Loss through reservoir	

The loss of Cs-137 was greater from cultivated watersheds than from uncultivated ones. On the basis of the results, the loss of radioactive fallout as determined for Cs-137 is very small (< 5%) from uncultivated watersheds. Some Cs-137 is lost through soil erosion, but much of this is trapped on sediments by the controlling reservoir.

Ricthie et al. (1971) studied the distribution of Cs-137 in a small watershed in northern Mississippi. The input of Cs-137 into the watershed from atmospheric fallout was estimated to be 159 nCi/m^2. This is the concentration measured in the Oak hickory woodlands in the watershed. These woodlands are in protected areas and have a thick layer of litter, a well-developed humus horizon and an A horizon indicating good infiltration and subsequently very little erosion or overland flow. Assuming a negligible loss of ^{137}Cs, the ^{137}Cs concentration in the oak-hickory woodland is therefore, indicative of the ^{137}Cs fallout over the watershed.

Concentrations of ^{137}Cs in the watershed ranged from 159 nCi/m^2 under the oak hickory sites to 8.9 nCi/m^2 for the eroded (no vegetation) sites.

Table 4 *Concentrations of ^{137}Cs under the land uses of Powerline watershed (from Ritchie et al., 1971)*

Land use	% of watershed	No.of Sites	^{137}Cs concentration (nCi/m^2)
Oak hickory	10	3	159.0
Pine	79	7	153.2
Grass (Pastures)	7	4	147.2
Eroded (no vegetation)	4	4	8.9
Average ^{137}Cs concentration in watershed			147.6

At all sites the concentration of Cs^{137} was greatest in the upper 2.5 cm of soil. Differences in concentrations of ^{137}Cs under the various land uses were correlated with the amount of erosion at each site. The pine areas vary in the amount of erosion but in general have only small areas of active erosion. The eroded areas are all subject to active erosion. The grass areas are overgrazed and have many areas of active erosion.

Table 5 *Concentrations of ^{137}Cs in the sediments of Powerline watershed (from Ritchie et al., 1971)*

Depositional Area	Number of sites	^{137}Cs concentrations (nCi/m^2)
Stream Channel & overbank deposits	1	2.3
Flood Post	4	207.3
Conservation Pool	4	412.2

The coarse textured stream channel and overbank sand deposits had the lowest ^{137}Cs concentration measured in the watershed. The coarse to medium textured sediments of the flood profile area had a ^{137}Cs concentration higher than one watershed average, but lower than the medium-to-fine textured sediments in the conservation pool of the reservoir. The distribution of ^{137}Cs in the different sediments is as expected since ^{137}Cs has been shown to be adsorbed by the fine-size fractions of soils.

Concentrations of ^{137}Cs in the conservation pool sediments was 2.8 times greater than the average ^{137}Cs concentration of the watershed when compared on a unit area basis. This indicates a movement of the ^{137}Cs from the watershed into the conservation pool. The ^{137}Cs probably moved largely on sediment particles eroded from the watershed, although a small percentage could have moved in solution in the runoff waters.

Table 6 *Model for the distribution of Cs-137 in soil and sediments of Powerline watershed (from Ritchie et al., 1975)*

Input from the atmosphere into reservoir and watershed

159 nCi/m²		input activity
125.85 ha		area
200.11 mCi		total activity

19.18mCi	151.48mCi	13.42mCi	7.66mCi	Input
Oak	Pine	Grass	Eroded	
159nCi/m²	153.2nCi/m²	147.2nCi/m²	8.9nCi/m²	Activity
12.06ha	95.27ha	8.44ha	4.92ha	Area
19.18mCi	145.95mCi	12.42mCi	0.43mCi	Total Activity

13.76mCi — Total Output

4.25mCi — Direct Input

Flood Pool

207.3nCi/m²	Activity
2.67ha	Area
5.53mCi	Total Activity

12.48mCi — Total Loss

4.12mCi — Direct Input

Conservation Pool

412.2nCi/m²	Activity
2.59ha	Area
10.68mCi	Total Activity

5.92mCi — Loss through reservoir

It can be seen from Table 6 that erosion processes are playing a major part in the redistribution of ^{137}Cs within the watershed. The trap efficiency (expressed as the ratio of the material retained to that entering a reservoir) of Powerline Reservoir is estimated to be 70 to 95% for all sediment particles. The trap efficiency calculated for Cs-137 in Powerline reservoir was 57%.

A study of the Longdendale valley and its reservoir indicated by Jones and Castle (1987), that the major part of the radioactive fallout was bound up in the grasslands on the valley sides. Analysis of samples taken from the River Etherow and Heyden Brook showed that very little radioactivity was being leached off the hills and that the water entering the reservoirs was diluting

the activity which had previously accumulated.

4. Soils

4.1 SOIL PLANT RELATIONSHIPS

When fallout is deposited a portion of it is intercepted by plants. The fraction of fallout thus adsorbed, and its residence time is of some importance in the cycling of radiocaesium within the soil. In a study of Alaskan vegetation Kodaira (1971) gave the following results. The residence half-time for ^{137}Cs determined from samples obtained in 1967 and later is not affected by current fallout levels, particularly for lichens and evergreen shrubs, which accumulate and retain fallout radioisotopes. The amount present in such plants are high due to the atmospheric weapons tests of 1961 and 1962. Peak fallout deposition occurred in 1963 and has decreased exponentially since then, so that in 1967 the deposition was less than 2% of the accumulated ^{137}Cs. The deposition of fallout from then recent tests conducted on mainland China is also considered to be negligible when compared with the total accumulated ^{137}Cs from the massive injections into the stratosphere by the 1961 and 1962 USA and USSR tests.

The residence time for ^{137}Cs in *Cladonia* sp. lichens collected in the higher elevations of Alaska ranged from 7.8 to 8.9 years with an average value of 8.1 years, compared with 17 years reported in Sweden. The residence time for lichens growing in areas of higher precipitation, and maritime moderated climates appeared shorter. This could be due to washout of Cs, or dilution because of increased growth rates.

From work on the cycling of caesium in Florida, Gamble (1973) stated that the evidence indicates that the radiocaesium in Florida ecosystems is tied up in non-exchangeable form with the organic fraction, and the abrupt disappearance of the activity in the mineral soil, plus low levels in the groundwater, confirm this observation and further demonstrate the efficiency of the cycle.

From studies of the effect of cropping on the release of caesium from soils Nishita (1982) found that the amount of exchangeable caesium remained roughly constant irrespective of the H ion concentration. This would tend to indicate that Cs-137 is relatively more strongly fixed than H^+ once it is on the exchange site. Under cropping conditions, Cs-137 in the soil solution and the H^+ ions from the plant roots, probably compete for the exchange sites as exchangeable and non-exchangeable K and other bases are absorbed by the roots. The apparent increased fixation of Cs-137 during cropping of the soil and the chemical evidence indicate that Cs-137 might compete effectively against H for the exchange sites in the soil.

The exchangeable Cs-137 decreased with cropping, but Cs-137 content of the plants remained constant for the first five crops and then increased. The Cs-137/K atom ratios increased as cropping progressed. The trends of the

ratio plots for clover tops and roots follow closely those of soil leachate. The maximum activity of Sr-90 and Cs-137 removed by a single crop was only 4.42 and 0.13% of dose respectively. The cumulative total uptakes of Sr-90 and Cs-137 for 9 croppings obtained over the period of 516 days were 23.69 and 0.72% respectively.

Significant amounts of Cs-137 were exchangeable (minimum of about 17% of dose) even after the maximum apparent increase in fixation which was about 7% of dose. Thus the increase of both the uptake by plants and the fixation was derived from the exchangeable form, as the competition offered by K diminished.

Since in solution culture experiments Cs has been shown to be absorbed as readily as Sr by several plants and more readily by others, the differential uptake of the two radioisotopes must be due primarily to the much lower availability of Cs-137 in the soil. The non-exchangeable fraction of Cs-137 in Vina loam was about 75% of dose while that of Sr-90 was about 7%. Because of the relatively strong fixation of Cs-137, the H^+ ion from the plant roots could not displace it from the soil exchange complex as readily as Sr-90.

4.2 SOIL CHEMISTRY

According to a number of investigators the adsorption energy of Cs^+ on clay minerals and on soil colloids generally exceeds that of any of the other alkali ions. Also, work with carrier free Cs-137 suggests that caesium may be irreversibly adsorbed in a manner possibly similar to the way potassium is fixed by many soil colloids and clay minerals.

It is observed by Schulz (1985) that total Cs^+ sorbed exceeds the CEC in the majority of soils and minerals. This effect is especially pronounced in Vina soil and vermiculite. It appears that those soils with the lowest soluble K fix the greatest amount of Cs. Also data allows for the possibility that Cs fixation is related to the vermiculite content of the samples. Drying effects a small increase in Cs-137 fixation in all materials except Hanford and Vina soils, where a reduction in fixation is noticed. The fixation of Cs-137 in soils in carrier-free (trace) amounts is an essentially different process from those predominant in the fixation of macro amounts of caesium, potassium and ammonium.

Leaching experiments, reported by Frissel and Pennders (1985) with ^{137}Cs in columns, showed that irreversibility plays an important role. Desorption occurred more slowly than adsorption. This phenomenon is known as fixation and occurs mainly in the presence of illitic clay minerals, the Cs ions are adsorbed rather specifically between the platelets of the illite crystal. Even traces of illite are sufficient to fix Cs and because all soils contain at least some illite, almost all fix Cs. The only exceptions are peat soils with a high K content. The release of fixed Cs is very difficult to predict.

It is theorised by Francis and Tamura (1971) that on entering the soil system Cs^+ is immediately adsorbed by any negative charges, mineral or organic. The movement on the mineral surface is then controlled by double layer diffusion processes; and as a consequence of caesium's low energy of hydration the

proportion of time it is present in the Stern layer is considerably greater than for other cations, e.g. Mg, Ca, Al.

Caesium has a high point charge and an ionic radius (0.169nm) conducive to adsorption at the hexagonal network at the interlayer surface of clay minerals. Also because it is easily dehydrated Cs has the ability to move more easily into the interlayer regions of 1.2 - 1.4 nm layer silicates than do hydrated cations. Once Cs^+ has entered such an interlayer region the probability of its exit is also much smaller than for the hydrated cations.

Trace distribution coefficients (kd values) for Cs, Sr and Co sorption were found by Routson *et al.* (1984) to be a function of sediment type (soil type) and of the solutions concentration of macro-ions including Na^+, K^+ and Ca^{2+} over wide macro-ion concentration ranges.

$$Kd = \frac{As\ /W}{Al\ /V}$$

As = equilibrium radioactivity sorbed on the solid
W = weight of solid phase (g)
Al = equilibrium radionuclivity of the solution
V = volume of solution in ml

In contrast to macro-ions, fission products in soil systems are generally in much lower concentrations ($\leq 10^{-5}$ M), so that their chemical competitive effects can be considered as negligible.

ln kd Cs*Na = 4.5 - 0.96 ln(nNa) - 0.078 $ln(Na)^2$	ln kd ~ .3-8
ln kd CS*K = 0.50 - 0.85 ln(K)	ln kd ~ 2-6
ln kd Cs*Ca = (no apparent relationship)	ln kd ~ 8-9

4.3 TRANSPORT

Due to the very strong fixation of caesium by soil minerals, little movement of caesium is expected down the soil profile. The process of movement within the soil has however been modelled by Frissel and Pennders (1985). They produced a process oriented model for strontium, and a semi-empirical one for caesium, plutonium, and americium. This compartmental model divided the soil into a series of layers, each of which had an input from the above layer and an output to the layer beneath, as well as a radioactive decay term. The residence time used for the quite successful model of caesium was 2years/cm. Thus by the time the caesium has penetrated to 10cm there has been substantial decay (half-life 30 years).

This work is corroborated by an investigation of the varying transport characteristics of abandoned and cultivated field plots by Cline and Rickard (1971). They found that after eight years, 70% of the added ^{137}Cs remained in the upper inch of untilled soil. In tilled soil, ^{137}Cs was concentrated at

the tillage layer depth of 5.6 in.

5. Groundwater

Effects of groundwater composition and redox potential (Eh) on radionuclide sorption and desorption of the geologic solids were studied by Barney (1984). He found that Na, K, and Ca in the groundwater, decrease sorption of Cs, Sr and Ra by ion exchange reactions.

Sorption and desorption isotherms were obtained for sorption of radionuclides under oxidising and reducing conditions. The Freundlich equation accurately describes most of these isotherms. Most radionuclides are apparently irreversibly sorbed on each of the geologic solids, since the slopes of sorption and desorption isotherms for a given radionuclide are different. This hysteresis effect is very large and will cause a significant delay in radionuclide transport. It therefore should be included in modelling radionuclide transport to accurately assess the isolation capabilities of a repository in basalt.

Precipitation and sorption on interbed materials are the principle reactions that retard radionuclide movement in the watersheds.

A frequently used simple sorption (or desorption) model is the empirical distribution coefficient, Kd. This quantity is simply the equilibrium concentration of sorbed radionuclide divided by the equilibrium concentration of radionuclide in solution. Values of Kd can be used to calculate a retardation factor. R, which is used in solute transport equations to predict radionuclide migration in groundwater. The calculations assume instantaneous sorption, a linear sorption isotherm, and single valued adsorption-desorption isotherms. These assumptions have been shown to be erroneous for solute sorption in several groundwater-soil systems. A more accurate description of radionuclide sorption is an isothermal equation such as the Freundlich equation.

$$S = K\ C^N$$

S = equilibrium concentration of sorbed radionuclide (moles/g)

C = equilibrium concentration in solution (moles/L)

K,N are empirical constants

The Freundlich was often found to accurately describe sorption and desorption of all radionuclides studied in the interbed-groundwater systems, except when precipitation of one radionuclide occurred.

Table 7 Freundlich constants for Radionuclide sorption (molar basis) on Sandstone(from Barney, 1984)

Radionuclide	Redox	Temp.	K	N
Caesium	Oxidising conditions	23°C	0.023	0.70
		60°C	0.0029	0.72
		85°C	0.0013	0.72

In each case, N was less for desorption and K was greater for desorption than adsorption. This chemical hysteresis will of course, affect radionuclide transport. Ignoring hysteresis effects could cause serious errors in predicting radionuclide movement.

KCl, NH_4 and NaCl reduce caesium sorption. K^+ and NH_4 appear to compete with Cs^+ for adsorption sites (cf.K^+ on mica like materials). In short, ions of similar size effectively compete with both Cs^+ and Sr^{e+} for sorption sites.

Over the range of groundwater component concentrations studied, metal complex formation with groundwater anions does not greatly affect sorption.

Results from Skagius and Neretnieks (1988) indicate that the diffusion of caesium and strontium (sorbing components) in rock material is caused by both pure diffusion and surface diffusion acting in parallel.

Industrial and low-level radioactive wastes at a National Reactor Testing Station (NRTS) in Idaho have been discharged to the Snake River Plain Aquifer since 1952. The aquifer is large and has a high transmissivity. This area was studied by Robertson and Barraclough (1973). He found that tritium and chloride had migrated as much as 8 km. The movement of cationic waste solutes, particularly Sr^{90} and Cs-137 has been significantly retarded owing to solution phenomena, particularly in exchange. Cs-137 has shown detectable migration in the aquifer and the Sr^{90} has migrated only about 2 km from a discharge well. The Sr-90 plume covers an area of only 4 km^2 of the aquifer.

Digital modelling techniques have been applied successfully to the analysis of this complex waste-transport system by numerical solution of the sampled equations of groundwater motion and mass transport. The model includes the effects of convective transport, flow divergence, two dimensional hydraulic dispersion, radioactive decay, and reversible sorption.

Of the significant longer-direct nuclides only tritium has entered the Snake River Plain Aquifer in detectable quantities. All the other nuclides are cationic and are removed from solution by sorption before reaching the water table. Cs-137 is even more affected by sorption than Sr^{90}. Although Cs-137 and Sr^{90} are produced and discharged in nearly equal quantities, Cs-137 has never been detected in any aquifer samples near the NRTS. Although there is

considerable dispersion laterally and longitudinally, there appears to be little vertical migration because of low vertical permeability.

6. Water Chemistry

6.1 DRINKING WATER QUALITY/MONITORING

There is very little data available on the concentrations of caesium in drinking water, although Crooks *et al.* (1959) gave some and Jones and Castle (1987) have carried out a number of analyses for the North West area. Further data for the North West is available in a North West Water planning document, Castle (1988a, b).

A summary of Jones and Castle (1987) who monitored radioactivity of the water cycle following the Chernobyl accident, is given below.

Analytical Methods

Samples were analysed for gross β-emission activity, a technique which does not allow the identification of the individual radionuclides.

Results

Analysis of Raw and Treated Water Samples

Of waters abstracted and treated for potable use and sampled for radioactivity, those from the River Dee at Huntingdon were found to have the highest significant levels of contamination. This was not surprising because the source of the River Dee and its extensive catchment is in North Wales where particularly heavy rain occurred at the time the cloud of radioactive material passed over that part of Britain from Chernobyl.

Analysis of Sludge Samples from Water Treatment Plants

Radionuclides were very effectively removed and concentrated in filter sludge cakes.

Sample		Activities (bq/kg) Cs 137	Cs 134	Treatment Process
Amfield	14.5.88	600	350	Coagulation with ion
Amfield	29.10.86	24	17	Coagulation with ion
Huntingdon	21.5.86	360	-	Coagulation with Al
Huntingdon	29.9.86	605	315	Ciagulation with Al
Kinder	21.5.86	-	-	Coagulation with Al
Watchgate	8.10.86	380	190	+ gravity filtration only

Analysis of Rainwater Samples

Most noticeable in the close correlation between the level of gross beta-activity in the rainfall and that in the water supply (Fig. 6). Other features of interest include the delay between the rise in the level of activity in rainfall following the bomb tests and that of treated water, and the surprisingly high level of Beta-activity recorded in the treated water following a sustained period of high gross ß activity in the rainfall.

Conclusion

An important side effect of the low levels of radioactivity occurring in surface water supplies is that freshwater fish, notably impounded brown trout in Ennerdale, Cumbria, accumulated levels of radioactivity up to 1000 Bq/kg.

Apart from distance, four factors limit the effect of the Chernobyl fallout:

(i) the period of heavily contaminated rainfall only lasted a few days;

(ii) a large proportion of the radioactivity was quickly bound up with the soil and grass;

(iii) radioactivity washed into water system was diluted considerably by the vast amounts of water already present;

(iv) a significant proportion of radioactivity was removed by water treatment processes.

7. Biology/Biota

7.1 MONITORING

The following is a resume of the monitoring programme and results following the Chernobyl reactor accident, as reported by Camplin *et al.* (1986).

Freshwater fish

The sampling programme focused on areas of high deposition, notably Cumbria, North Wales and the Isle of Man. Brown and rainbow trout account for the majority of samples but a variety of other species were analysed, including char, sea trout, salmon, perch and eels. Iodine-131 was found in several of the early samples, but at a maximum of less than 7 Bq kg^{-1}. Initially, little or no other activity was found in freshwater fish, other than low concentrations of caesium-134 and -137. More recently and as expected, higher concentrations of caesium were detected, notably in brown trout from impounded waters (up to 1,000 Bq kg^{-1}), but also in perch and other, mainly sport, fish (up to 2,000 Bq kg^{-1}) which are not eaten in large amounts and thus, compared with trout, have little or no significance as a source of potential radiation exposure. The higher concentrations are confined to soft water areas but are not general even there; the measurements cover a wide range with only a small proportion of fish showing high values.

This slowly developing caesium situation in certain freshwater fish is most likely due to a combination of mechanisms including delay in run off, turnover time of the water body, build-up via the food chain and slow turnover of these radionuclides in fish, the biological half-time of caesium in fish being about 100 days (Hewett and Jefferies, 1976). However, probably the most significant factor is that high deposition occurred in very soft water areas in which it has already been established (Preston *et al.* 1968) that very high concentration factors occur for caesium.

Maximum Radioactivity Concentration

In Trawsfynydd, the only lake for which there appears to be data spanning at least 6 readings for brown trout and rainbow trout the maximum in radioactivity concentration appeared to be between the 7 and 14 June, although a very high figure is also obtained for 7 August. However, this later peak may have been due to later deposition from a Chernobyl contaminated air pocket in the region, or discharge from Trawsfynydd as suggested by the increase in Cs-137:Cs-134 ratios in the water.

N.B. Caesium 134 : Caesium 137 ratios initially less than two for Chernobyl, 13-15 for BNFL Sellafield discharges

The Chernobyl reactor accident and its impact on the aquatic environment (marine and freshwater) has also been studied by MAFF (1987). They report monitoring of aquatic foodstuffs in the light of previous experience with weapons testing activity. Although activity levels in aquatic foodstuffs did not exceed the limits of safety, the highest activities occurred very shortly after deposition (within a day or two), partly due to runoff from adjacent land.

Although concentrations rose very sharply, the peak values were not sustained and declined rapidly.

In the early phase very little activity was found in freshwater fish, though, as time went on concentrations of Cs-134 and Cs-137 began to increase. This only occurred in indigenous fish in the few areas of the country that had experienced high rates of deposition.

Research related to the discharge of radionuclides in coastal waters being undertaken by MAFF, include the following. Further information on each of these topics is given in MAFF (1987).

(i) The Association of radionuclides with settled sediments

(ii) The incorporation of radionuclides into the sea bed

Bioturbation has been found to cause spatial heterogeneity in distribution of radionuclides.

(iii) The Geochemistry of interstitial water

The sediments often have a lower radio potential than the overlying water. This can result in both plutonium 239 + 240 and Americium 241 being present in the reduced form. The reduced form of both radionuclides has a higher sediment distribution coefficient than the higher valence states, and leads to their preferential retention.

Bioturbation could cause significant changes in speciation as a result of redistribution between the oxic sediment and the anoxic area at greater depths.

(iv) The Irish Sea model

The water circulation pattern for the Irish Sea may be observed or modelled. Observations of ^{137}Cs distributions have been made extensively. The source term, the annual (and monthly) discharges of ^{137}Cs from Sellafield, is known and the nuclide is essentially conservative with the water, undergoing relatively little scavenging by particulates and transfer to the sea bed.

(v) Eastern Irish Sea sediment dynamics study

Dissolved contaminants, such as ^{137}Cs can be measured to provide an average picture of how water movements disperse discharges.

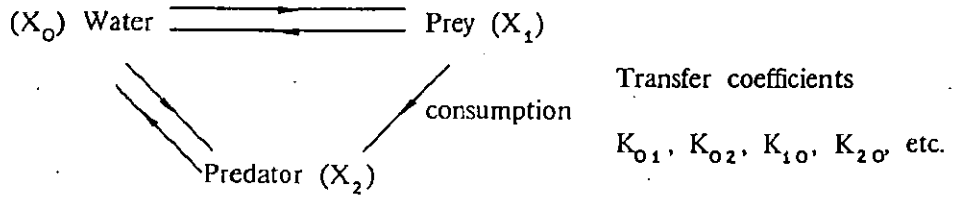
Concentrations of Cs-134 and -137 were initially very low in freshwater fish

everywhere, and this situation has remained in lowland areas where deposition was low and generally where the water is hard. Within a few weeks, Cs concentrations in fish began to increase. These were confined to areas of soft freshwater that had also experienced higher rates of deposition, notably North Wales, Cumbria, parts of Scotland and the Isle of Man, though even there they were not general with only a small proportion of fish showing the higher values. The level of Cs in farmed and stocked trout, mainly of the rainbow variety, has remained low; these are the principle sources of trout that are eaten. The highest concentrations have been in indigenous fish in the wild, e.g., Brown trout (up to about 1000 Bq kg^{-1}) and perch, pike and other mainly sport fish (up to about 2000 Bq kg^{-1}) that are not eaten in sufficient amounts, compared with trout, to constitute a potentially significant source of radiation exposure. Some of the highest rates have been found in Lake Trawsfynydd though much of the activity there is due to the discharges from the CEGB nuclear power station.

7.2 FOOD CHAIN ANALYSIS

The concentration process of contaminating radionuclides along a food chain undoubtedly plays an important role in determining the overall concentration in fish. In Aoyama (1978a) the effect of dietary intensity, feeding interval and biological dilution due to growth is investigated and a model of a food chain is presented in another paper (Aoyama *et al.*, 1978b). The results of this work suggests the concentration of metal in predator fish increased with ration size, was unaffected by feeding interval and that biological dilution of heavy metal concentration takes place in the growth period.

The model presented takes as its basis the simple 3-compartment system of water, prey and predator.



The coefficient of most interest, K_{12} can be expressed as

$$K_{12} = \theta M_1/M_2$$

θ = Assimilation rate

M_1 = Ration size

M_2 = Predator size

M_1 is affected by the population density of food, the distribution of food, and the ecological structure of the food organism population. A mathematical

treatment allows the combination of these factors into one, the food concentration factor. Furthermore, factors to account for the effect of growth were included.

In a study of the paths to human dosage in the Hudson River Estuary, Linslata *et al.* (1986) give figures consistent with previously reported effective half-lives for Cs-137 clearance from estuarine fish of between 100 and 200 days. Linslata *et al.* also report marked inter- and intra-species variation in fish caesium concentrations, dependent on migratory and feeding habits, in particular the seasonality of behaviour and whether or not the fish feed on the bottom.

Radiocaesium analyses have also been made for vegetation along a contaminated stream length by Briese *et al.* (1975). These tend to indicate that leaf concentrations are higher in plants where the rate of stream flow decreased because of impoundments or impediments. The distribution of concentrations were such though that one plant species could not be used to predict concentrations in another.

8. Modelling

A model has been developed by Wrenn *et al.* (1971) to predict the average amount of Cs-137 in fish, which depends upon the average concentration of radiocaesium in water and in sediments.

Although the ^{137}Cs content of the Hudson River has dropped tenfold since 1964, the average content in fish and sediments has remained relatively constant. This implies that the Cs-137 content of the resident fish in the river has depended primarily upon the cumulative deposit in the sediments. The major source of this ^{137}Cs has been fallout, although in a limited section of river an elevation of ^{137}Cs in sediments from the Indian Point Nuclear Plant can be seen. In order of decreasing content, ^{137}Cs was found in sediments, aquatic plants and fish roughly in the ratios of 30:1:1.

The average content of radiocaesium in fish has shown surprisingly little variation from year to year, from a minimum of 20 pCi/kg in 1967 to a maximum of 56 in 1969. Cs-137 has been measured in 16 species of fish and is generally an order or magnitude lower than ^{137}Cs content of fish from lakes at this latitude. In relative terms the highest concentrations of Cs-137 are found in bottom feeding species, intermediate levels in plant-eating species and the lowest levels in anadromous fish. Apart from anadromous fish, the range of variability is about a factor of 3.

The environmental characteristics which influence the expected concentrations to be found in fish are the water chemistry, the cumulative deposition characteristic of a given region, the accumulation of ^{137}Cs in bottom sediments, the species and feeding habits of fish, the temperature and the D0 content.

In the Hudson River the Cs-137 content of fish depends upon both the

cumulative accumulation in sediments and the mean concentration in water. The ^{137}Cs content of fish appears to be independent of the concentration of K in water in the range studied between 2 and 30 mg/litre
The major portion of ^{137}Cs present in the estuary is associated with the sediments. Generally ^{137}Cs does not appear to increase with trophic level. The variability among indigenous fish is only about a factor of 2/3.

MODEL FOR TRANSPORT OF RADIONUCLIDES FROM EFFLUENT RELEASES

The joint CEC, NRPB document published by the CEC in 1979 presents a model for the dispersion and transport of radionuclides released as effluents from nuclear power plants.

It encompasses three main processes -

1. Physical movement and dispersion of water masses.
2. Interaction of radionuclides with suspended matter and bed sediment.
3. Radioactive decay.

The model which is developed is intended to represent time dependent behaviour but relies heavily on equilibrium concentration data to estimate the transfer of activity.

The model assumes that radioactive effluents are diluted in the river flow and that activity is adsorbed on the sediments to an extent depending on the radionuclide. Activity is transported downstream by the river flow and by the slower movement of contaminated bed sediments. After the termination of the discharge the activity in water is taken as zero, because it has been assumed that there is no desorption of activity adsorbed on sediments.

The concentration, C_w per unit discharge rate, in the river at a distance x from the point of release is given by -

$$C_w = \frac{1}{q} e^{-kx}$$

where,

k = factor dependent on river and radionuclide (m^{-1})
q = river flow rate (m^3/s^{-1})

For a given nuclide the value of k depends on its half-life, the river water velocity, and the extent of sedimentation effects

$$k = \frac{\lambda}{W} + K^1 (m^{-1})$$

where,

λ = radioactive decay constant (s^{-1})
W = river velocity ($m\ s^{-1}$)
K^1 = depletion factor for sedimentation (m^{-1})

The concentration $C_w = C_f + C_m M$ (Bq^{-3} per Bq $s^{-1/6}$)

where,

M = mass of suspended sediment (t m^{-3})

C_m = activity on suspended sediments (per unit release rate)

C_f = activity dissolved in water (per unit release rate)

The activity on the suspended sediments is assumed to be related to the activity in the river water

$$C_m = K_d \, C_f$$

where,

K_d = distribution coefficient of nuclides on sediment (Bq t^{-1} per Bq m^{-3})

Substituting above;

$$C_f = \frac{C_w}{1+K_d M} = \frac{1}{q} \cdot \frac{e^{-kx}}{(1+K_d M)} \quad (Bq\ m^{-3} \text{ per } Bq\ s^{-1})$$

$$\Rightarrow \quad C_m = \frac{K_d}{q} \cdot \frac{e^{-kx}}{(1+K_d M)} \quad (Bq\ t^{-3} \text{ per } Bq\ s^{-1})$$

Sediment activity can be transported by flow under the influence of the shear force exerted by the river flow and to a lesser extent gravit, such that the average velocity of the river bed sediments (V) is defined;

$$V = \alpha \sqrt{h \times J} \quad (ms^{-1})$$

where,

h = depth of water (m)
J = slope of river bed
α = constant ($m^{1/2}\ s^{-1}$)

A full mathematical derivation of the sediment transport equation is given in the appendix to this section (4) of the CEC publication.

The activity, Q_s in the sediments is a function of time, and distance from the point of release. During a period of release the profile is given by;

$$Q_s(x, t) = \frac{k^1 \, Q_w}{KV - \lambda} \cdot [e^{(KV-\lambda)t} - 1]$$

where,

$Q_w(x)$ = activity in water (Bq)

$Q_s(x)$ = activity in sediments (Bq m^{-1} per Bq s^{-1})

This model could be adapted to simulate transport in a complex system by stepwise division of the river into sections. The model has been successfully applied to the Rhone and the Loire using the following freshwater concentration factors for fish;

	Sediment	Fish
Freshwater concentration factors - Cs-137	30,000	1,000

Concentration factors here are the ratio of quantity per unit weight of the material concerned, and the quantity of activity per unit volume of filtered water, (Bq $tonne^{-1}$ per Bq m^{-3}). These are based on dry weight of sediments and wet weight of edible part of other materials.

PHYSICAL TRANSPORT MODEL OF CAESIUM THROUGH A CATCHMENT

A very useful model has been produced by Carlsson (1978). It is based on several simplifying assumptions, but highlights the key processes of caesium transport within and through a catchment. Like the other models so far discussed it delineates the components which contain caesium. These are:

> soil, vegetation, water of the lake, suspended sediment, bottom sediment and biological organisms.

Transfer between these compartments is then described by simple linear differential equations, and so the change of the ^{137}Cs activity in water with time can be exprezzed as;

$$\frac{dA}{dt} = U - p \times U - (\lambda + k) \times A$$

$$k = \text{outflow constant} = \frac{Q}{V} = \frac{\text{volumetric outflow rate}}{\text{volume of lake}} \left(\frac{m^3/yr^{-1}}{m^3}\right)$$

λ = physical decay constant (yr^{-1})

p = proportion of activity deposited to sediment

A = total activity in water (nCi)

U = rate of activity input (nCi/yr)

The rate of input of acting (U) can be ascribed to three processes -

$$U = U_1 + U_2 + U_3$$

$$U_1 = Y_S \times F_a$$

$$U_2 = K_1 \times Y \times F_a$$

$$U_3 = k_2 \times Y \times F_C$$

U_1 = direct deposition to lake surface

U_2 = wash-off of freshly deposited fallout

U_3 = wash-off of accumulated deposit

Y_S = area of the lake (m^2)

Y = area of catchment drainage area (m^2)

F_a = activity deposition rate ($nCi/m^2/yr$)

F_C = cumulative deposition (nCi,m^2)

K_1 = fraction of fresh fallout in the drainage area which is transferred to the lake

K_2 = fraction per unit of time of the cumulative deposition in the drainage area which is transferred to the lake (yr^{-1})

The cumulative deposition (F_c) can be calculated from -

$$\frac{d F_C}{dt} = (1 - K_1) \times F_a - (\lambda + K_2) \times F_C$$

Carlsson suggests that K_2 is proportional to the runoff of surface water from the drainage area to the lake. This means that K_2 has a seasonal variation with highest values during spring and autumn when the precipitation is large and the evaporation is small.

Rewriting the above equation:

$$\frac{dA}{dt} = (1 - p) \times (Y_S \times F_a + K_1 \times Y \times F_a + K_2 \times Y \times F_C) - (\lambda + K)A$$

p, Y_S, Y, λ and K_1 are constants

F_a, F_C, K and K_2 are functions of time.

Carlsson suggests treating the time dependent functions as stepwise functions,

with an interval of 0.2/yr.

If V is taken as being constant then C_w = activity concentration in Water = A/V

and so solving the differential equation gives

$$C_w = C_{w\theta} \times e^{-(\lambda+K)\ \Delta t} + \frac{(1-p) \times (Y_S \times F_a + K_1 \times Y \times F_a + K_2 \times Y \times F_c)}{(V\ (\lambda + K} \times (1 - e^{-(\lambda+K)\Delta t})$$

$C_{w\theta}$ = activity concentrated at time t

C_w = activity concentrated at time t + Δt

and for the sediment

$$\frac{dS}{dt} = P \times U - \lambda \times S$$

which similarly gives

$$S = S_0 \times e^{-\lambda\Delta t} + \frac{p \times (T_S \times F_a + K_1 \times Y \times F_a + K_2 \times Y \times F_c}{\lambda} \times (1 - e^{-\lambda\Delta t})$$

By a least square fit to the data Carlsson produces figures of

k_1 = 0.019

k_2 = 0.0056 yr^{-1}

p = 0.376

These figures yield the general conclusion that a large amount of the Cs-137 introduced into the lake is stored in the bottom sediment. The calculated rates of elimination of Cs-137 from the drainage area show that the radionuclide is strongly bound to soil and vegetation. The redistribution of Cs-137 between the different compartments of the watershed is small. The loss of Cs-137 is small and the main process is the physical decay.

A SPECIFIC ACTIVITY AND CONCENTRATION MODEL APPLIED TO CAESIUM IN AN OLIGOTROPHIC LAKE

Vanderploeg *et al.* (1975) has produced a linear systems analysis model derived to simulate the time-dependent dynamics of specific activity and concentration of radioactivity in aquatic systems.

The basic dynamic viewpoint is that stable element and radionuclide flow can be treated by first order linear differential equations and that the dynamics of specific activity follow along naturally. *Let,*

Z_i = mass of stable element in compartment i (g)

$a_{ij} Z_j$ = rate at which stable element moves from compartment j to compartment i (g/day)

$\gamma_i Z_i$ = rate at which stable element enters compartment i from the surroundings external to the system (g/day)

R_i = rate at which stable element enters compartment i from the surroundings external to the system (g/day)

Then;

$$\frac{d Z_i}{dt} = \sum_j{}' a_{ij} Z_j - (\gamma_i + \sum_j{}' a_{ji}) Z_i + R_i$$

where the primed summation symbol $\sum_j{}'$ is taken to mean the sum over all j except i.

The radionuclide (Cs-137) is presumed to move between compartments following the same laws as the corresponding stable element, so X_i, the amount of radionuclide in compartment i changes according to the relation;

$$\frac{d X_i}{dt} = \sum_j{}' a_{ij} X_j - (\gamma_i + \lambda + \sum_j{}' a_{ji}) X_i + k_i$$

where

k_i = rate at which radioactive element enters compartment i from surroundings external to the system (μCi day^{-1})

λ = physical decay constant of the nuclide (day^{-1})

Specific activity is defined as

$$S_i = \frac{X_i}{Z_i}$$

and transfer coefficients as

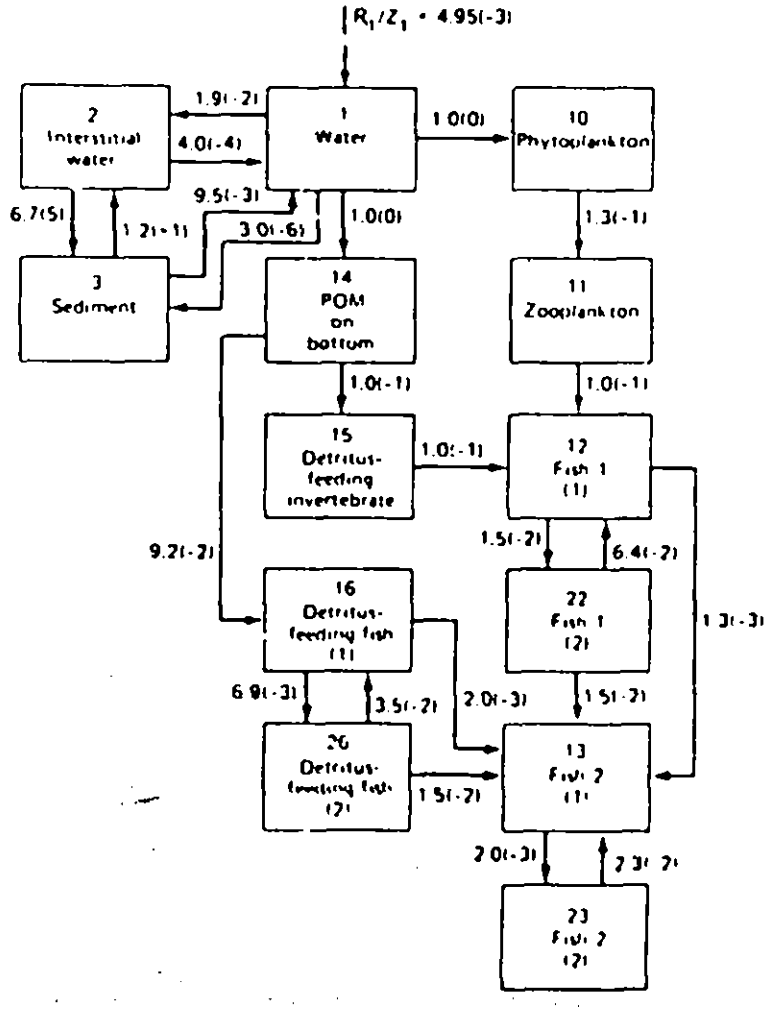
$$\alpha_{ij} = \frac{Z_j}{Z_i}$$

the specific activity relation is obtained as;

$$\frac{d\ S_i}{dt} = \sum_j^i \alpha_{ij} (S_{ij} - S_i) - \lambda\ S_i + \left(\frac{k_i}{Z_j} - \frac{R_i}{Z_i}\ S_j \right)$$

Essentially then. α_{ij} is the flow of stable element (g/day) into compartment i from compartment j divided by the amount of stable element in compartment i. Thus α_{ij} is in units of day^{-1}. The structure of the lake-ecosystem model is shown in Figure 2.

Figure 2 *Control diagram and transfer coefficients for simulation of specific activity flow in a hypothetical oligotrophic lake. Values of a_{ij} are indicated on arrows. Numbers are given in a form such that 1.9 (-2) is read as 1.9 x 1^{-2}.*



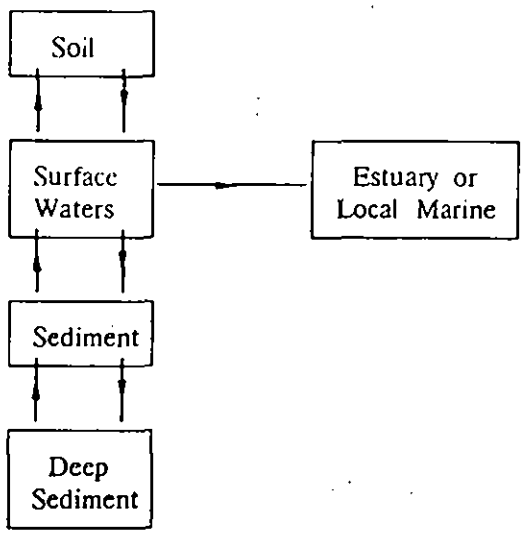
MODELS AND DATA TO PREDICT RADIONUCLIDE CONCENTRATIONS IN FRESHWATER FISH FOLLOWING ATMOSPHERIC DEPOSITION: SUMMARY OF PROGRESS TO MID-1988.

This report published by the NRPB and the MAFF Directorate of fisheries Research presents a simple model of caesium transfer into lake water (Ennerdale, Loweswater) from atmospheric deposition and thence into fish.

BIOS: A MODEL TO PREDICT RADIONUCLIDE TRANSFER AND DOES TO MAN FOLLOWING RELEASES FROM GEOLOGICAL REPOSITORIES FOR RADIOACTIVE WASTE

The conceptual basis of the BIOS model is shown in Figure 3.

Figure 3 *Schematic representation of freshwater aspect of biospheric transport model.*



Radionuclides deposited in a river system will interact with sediments and may also be transported to adjacent land areas as a result of irrigation practices, flooding, or sediment movement. Because of the difficulty in obtaining the site specific data required to calibrate the more complex advection-dispersion equation models, a simple compartment model of the river flow is used. The transfer coefficient from one freshwater compartment to the next is simply the volumetric flow (m^3y^{-1}) in that body divided by its volume.

The interaction of radionuclides and river sediments is modelled by use of Schaeffer's model detailed in CEC/NRPB (1979). Within each compartment representing a river section, the rate of removal of a radionuclide from the water column to the bed sediments is given by,

$$dI_w/dt = - V_w K I_w$$

where,

L_w = quantity of the radionuclide in the water column (atoms)

t = time (y)

V_w = velocity of river water in that section (m y^{-1})

K = depletion factor for sedimentation (m^{-1}) .

for caesium which interacts strongly $K \approx 10^{-5}$ m^{-1}.

N.B. As K is empirically derived both adsorption and desorption are accounted for.

Transport downstream is taken as being on both suspended sediment and in dissolved form. Partitioning is determined by the freshwater sediment distribution coefficient and the suspended sediment load according to the relation,

$$F_w = 1/(1 + K_d \alpha)$$

where,

α = suspended sediment load (t m^{-3})

K_d = freshwater sediment distribution coefficient which is element dependent (m^3 t^{-1})

F_w = friction of the quantity of a radionuclide in the water column which is in solution.

Values of α are reported to range from 10^{-4} t0 10^{-6} t m^{-3} and are strongly dependent upon the volumetric flow of the river. The coefficient used to describe the transport of radionuclides in bed sediment from one river to the next is given by,

$$K_{ij} = V_i/L_i$$

where

K_{ij} = transfer coefficient from section i to section j (y^{-1})

V_i = velocity of bed sediment in section i (m y^{-1}),

and L_i = length of section i (m).

In the case of radionuclides and lake sediments, a particle-scavenging model is employed. Radionuclides are partitioned according to;

$$f_w = 1/(1 + k_d\alpha)$$

and the component transported to the lake bed is given by the sedimentation rate, which can vary with the suspended sediment load of the inflow to the lake. The various components with their integral modelling procedures can then be interconnected in a manner applicable to any particular catchment.

MODELLING RADIOTRACES IN SEDIMENTS: COMPARISON WITH OBSERVATIONS IN LAKES HURON AND MICHIGAN

Christensen and Bhunia (1986) produced a comprehensive model for the activity of radionuclides in sediments. The model is based on the advection-diffusion equations for sediment solids and a radioactive tracer. The model is complex and generally an improvement on previous models, incorporating bioturbation as well as diffusion.

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The demand for long-term scientific capabilities concerning the resources of the land and its freshwaters is rising sharply as the power of man to change his environment is growing, and with it the scale of his impact. Comprehensive research facilities (laboratories, field studies, computer modelling, instrumentation, remote sensing) are needed to provide solutions to the challenging problems of the modern world in its concern for appropriate and sympathetic management of the fragile systems of the land's surface.

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